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DEFECTS IN HYPERPURE Fe-BASED ALLOYS CREATED BY 3MeV e⁻-IRRADIATION

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ABSTRACT:

Information about vacancy defects created in RPV(Reactor Pressure Vessels) steels after neutron irradiations are obtained via a simulation:

The RPV steels are simulated by a series of high purity Fe-based alloys representing the industrial alloy composition.

The neutron irradiation is simulated by a 3MeV electron irradiation.

Vacancy defects characteristics are obtained by positron lifetime techniques. Irradiations are made at 150°C or 288°C, with a dose of $4*10^{19}$ e-/cm^2, and followed by isochronal annealing in the range 20 -500°C.

The observed vacancy defects are single trapped vacancies and small vacancy clusters, the size of which being lower than 10 empty atomic volumes(Vacancy clusters containing more than 50 empty atomic volumes were never found). A large recovery step is observed between 200 and 400°C, after 150°C irradiation and attributed to vacancy-impurity detrapping, and also, vacancy cluster evaporation. The influence of C, Cu and Mo are presented.

These results are in agreement with a model supposing, in pure Fe, single vacancy migration at -50°C and vacancy-impurity detrapping at 200°C.

INTRODUCTION:

The irradiation damage play an important role in the security of nuclear reactors. Several studies were made about the neutron irradiation enhanced diffusion, the impurity agglomeration and the vacancy clustering[1,2,3,4,5].

This work is a part of a general study of vacancy type defects in Fe-based alloys using positron lifetime techniques:

In pure Fe, it was shown that single vacancy migrates at 220K[6];

In FeNiCr austenitic alloys, vacancy clusters overcoming 1000 empty atomic volumes were evidenced after an electron irradiation[7,8];

In EDF(Electricité De France) industrial ferritic steels, the size of observed vacancy clusters never overcome 50 empty atomic volumes under normal neutron or electron irradiations[9,10].

EXPERIMENT:

The Fe-based alloys were melted in CENG with hyperpure Fe containing less than 10appm of C+N[11,12], and Johnson Matthey Cu, Mn and Mo. Alloys of Fe, FeCu(0.1%), FeCu(0.1%)C (0.002, 0.004, 0.01%), FeCu(0.1%)Mo(0.5, 1, 2%) and FeCu(0.1%)Mn(1.2%) were elaborated. Before irradiation, they were annealed under 10^{-6} Torr, during 8h at 623°C(900K), and slowly cooled.

The samples were irradiated at 150°C and 288°C with 3MeV electrons to a standard dose of $4*10^{19}$ e-/cm^2, obtained in 72h using the Van de Graaff accelerator of CENG/SP2M/LPI. These irradiation conditions were chosen because:

*150°C is below dissociation temperature of vacancy-carbon complexes given by Arndt[13];

*288°C is the temperature of cold leg of the pressure vessel of French reactors;

*The 288°C standard electron dose appears convenient to represent a 13 years neutron irradiation in RPV steels.

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For a comparison between irradiation- and coldwork-vacancy defects, a set of specimens was heavily cold-worked with a cold rolling reducing the specimen thickness of 80%.

The isochronal annealing(ΔT=50K, Δt=30min.) were carried out under vacuum until 423°C for irradiated samples and 723°C for coldworked samples. At each step of the isochronal annealing, a positron lifetime spectrum is obtained. A spectrum of 10^6 events is obtained with a spectrometer having a FWHM 240ps in operating conditions. Positrons come from a ~20µCi ^{22}Na source sandwiched by Ni foils. Experimental conditions and one or two lifetime analysis are described elsewhere[8].

RESULTS :

Figures 1 ~ 4 presents the evolutions of the positron average lifetime in different conditions. The error bars on the average lifetime are ± 1ps and are not represented on the Figures. The variation of long or short components and their intensity are not presented here and will be published rapidly[14].

Figure 1 presents the evolution of the positron lifetime after a standard 150°C irradiation as a function of the annealing temperature, in pure Fe and in three Fe-based alloys. Values of the lifetime before irradiation are around 120ps: they are indicated above the point 280K. After a 150°C irradiation, the lifetime increases, especially for FeCu(0.1%) alloy(~220ps). In pure Fe, the lifetime increase is very small, 3%.

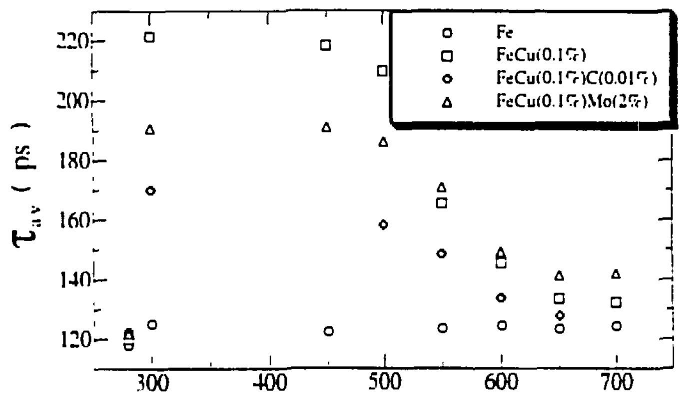

Figure 1 The dependence of lifetime on annealing temperature in 150°C irradiation alloys

In the three investigated alloys, a very large recovery step is found between 200°C and 400°C. It look too large to be attributed to a unique recovery process. In FeCu(0.1%)and in FeCu(0.1%)Mo(2%), the recovery is not terminated after an annealing at 427°C. This recovery step can't be detected in pure irradiated Fe.

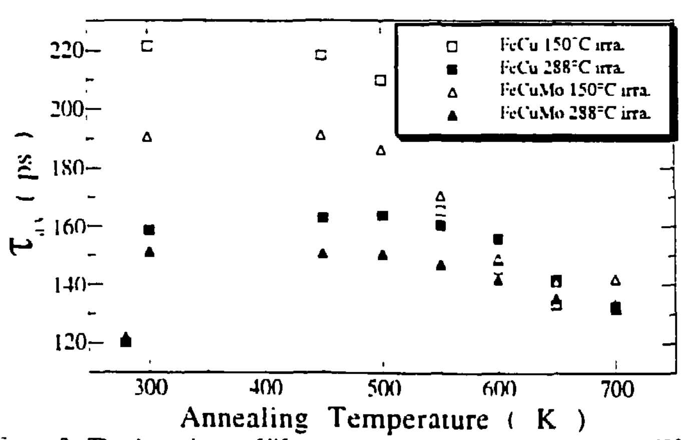

Figure 2 The dependence of lifetime on the annealing temperature in 150°C and 288°C irradiation

Figure 2 presents the results of FeCu(0.1%) and FeCu(0.1%)Mo (2%) after 288°C irradiation. Results after a 150°C for the same alloys are presented as a comparison. For the FeCu (0.1%) alloy, we note the crossing of the 150°C and the 288°C-cuvre, attesting an higher lifetime after a 288°C irradiation than after a 150°C followed by a convenient annealing.

On figure 3, the results of an heavy coldwork is presented. The average lifetime is increased in all alloys, and also in pure Fe. Here also, the maximum effect is obtained in FeCu(0.1%). Two recovery steps are observed in all

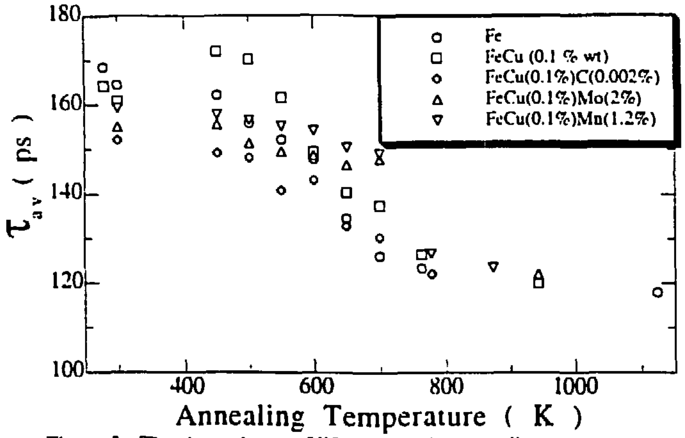

Figure 3 The dependence of lifetime on the annealing temperature in coldwork alloys

specimens. The temperature of the second step appears impurity dependent and is shifted towards high temperatures if Mo, Mn are present.

Figure 4 presents the variations of the average lifetime in FeCu(0.1%)C(x) and FeCu(0.1%)Mo(x) as a function of the x_C wt%, or x_{Mo} wt%. Before irradiation, the lifetime is constant and about 120ps, not far from the value of pure Fe. After a 288°C irradiation, the lifetime appears very sensitive of the carbon content, because τ_{av} is reduced from 160ps to 128ps by 0.002 wt% of carbon. Molybdenum has the same effect but a concentration of 0.4 wt% is need to obtain the same effect.

DISCUSSION:

Pure Fe: After an irradiation at 150°C, the τ_{av} is very near of the value observed in well annealed, non irradiated iron. This result indicates that no vacancy defects are retained in the hyperpure iron after 150°C irradiation. This result is agreement with the model previously presented: The 150°C irradiation creates single vacancies with a 0.55ev migration energy[6]. So, they migrate very quickly at 150°C, without having a chance to meet another vacancy and forming a polyvacancy. They disappear in dislocation or grain boundaries, without impurity-trapping because the iron is very pure.

On the contrary, in a 20°C coldwork, mono and polyvacancies are simultaneously formed(and also polyinterstitials, not detected here). The evolution of the long components, well above the value of the dislocation, indicates that polyvacancies are present. They are stable till 200°C and then anneal out in a recovery step. This is in accordance with the previous experiments of coldwork and/or irradiation of hyperpure iron at 20K[6] or at 27°C[15].

Fe(0.1%)Cu: After 150°C irradiation, a high τ_{av} is observed. Two components analysis reveals the existence of small vacancy clusters, attesting the property of Cu to cluster vacancies in three dimensional voids of 5 to 10 empty atomic sites, in concentration of about $5*10^{-8}$ (vacancy/atom). Such vacancy clusters grow in size during annealing, then evaporate above 200°C.

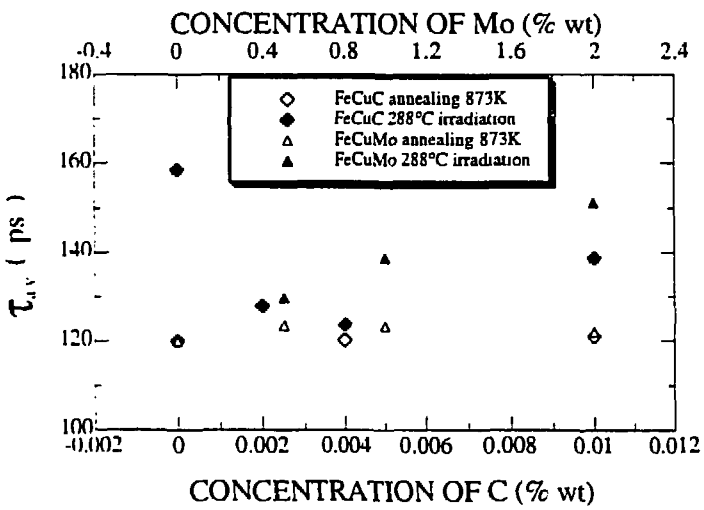

Figure 4 The dependence of lifetime on the concentration of C, Mo

After a 288°C irradiation, different defects are formed, involving 2 or 3 vacancies, in concentration about 10^{-7}(vacancy/atom) , which are stable till 350°C.

After a 20°C coldwork, the big increase of the lifetime confirms the property of Cu to collapse small vacancy aggregates.

Fe(0.1%)Cu(0.002, 0.004 and 0.01%)C: It is interesting to note that a very few quantity of carbon(0.002 wt% in case of figure 3 and 0.01 wt% in case of figure 1) reduces the value of the lifetime. The same effects appears on figure 4, after a 288°C irradiation. This confirms the high purity of the CENG iron.

Fe(0.1%)Cu(0.5, 1 and 2%)Mo: In presence of Mo, vacancy defects anneal out at higher temperature, +100°C for 2% of Mo. The impurity Mn seems to have the same behaviour.

On figure 4 is shown the property of Mo to decrease τ_{av}. Before concluding that Mo acts as carbon, with an efficiency reduced by a factor 200, we have to confirm the carbon concentration of our Molybdenum.

Recovery Step 200°C - 400°C: This step is too large to correspond to a unique mechanism. On figure 1, this recovery step is found for any irradiated alloys except for irradiated pure Fe. Figure 3 show that a similar step is observed in all specimens after 20°C coldwork, including pure Fe.

Below this step, trapped single vacancies are detected by a two lifetime analysis(with the exception of FeCu(0.1%), where trapped polyvacancies are detected). Above this step, there are absent. However, small vacancy clusters survive. As suggested by Arndt[13], this step corresponds certainly to detrapping of vacancies from impurity(essentially Carbon). Vacancy cluster evaporation occurs probably just above this range of temperature.

CONCLUSIONS:

These experiments are in agreement with the hypothesis of the isolated single vacancy migration at -50°C and the single vacancy-impurity dissociation above 200°C.

The recovery step observed above 200°C is too large to be attributed to only one process. In our interpretation it corresponds to single or poly vacancy-impurity dissociations in its first part and vacancy-clusters disparition in its final part.

Carbon trap single vacancy, copper creates small vacancy clusters and Mo, Mn stabilize strongly irradiation or coldwork vacancy defects.

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